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Short communication. The antifeedant activity of natural plant products towards the larvae of *Spodoptera littoralis*

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Abstract

Worms are becoming difficult pests to control since many insecticides are now forbidden and the present alternatives are insufficiently effective. Antifeedants of plant origin may have potential against such pests. In this work, several diterpenoids, flavones and coumarins extracted from plants were assayed as antifeedant agents against *Spodoptera littoralis* larvae. Among these compounds, four diterpenes (eriocephalin, salviacoccin, aethiopinone and oxocandesalvone) and four coumarins (oxypeucedanin, xanthotoxin, isoimperatorin and prangol) showed significant activity, whereas all the flavones tested were inactive. Some aspects of the structure-activity relationships of these compounds are briefly discussed.

Additional key words: coumarins, diterpenes, flavones, higher plants, secondary metabolites.

Resumen

Comunicación corta. Actividad antialimentaria de fitoquímicos naturales en larvas de *Spodoptera littoralis*

En la actualidad, debido a que se están prohibiendo muchos insecticidas y a que las alternativas disponibles no son completamente eficaces, las orugas están resurgiendo como plagas difíciles de controlar. Los compuestos antialimentarios de origen vegetal se consideran con posibilidades para desarrollar nuevos agroquímicos. En este trabajo, diversos diterpenoides, flavonas y cumarinas procedentes de plantas han sido ensayados como antialimentarios frente a larvas de *Spodoptera littoralis*. Tres diterpenos (eriocefalina, salviacoccina, aetiopinona y oxocandesalvona) y cuatro cumarinas (oxipeucedanina, xantotoxina, isoimperatorina y prangol) presentaron una actividad significativa, mientras que las flavonas fueron inactivas. Se discuten brevemente algunos aspectos de la relación estructura-actividad.

Palabras clave adicionales: cumarinas, diterpenos, flavonas, metabolitos secundarios, plantas superiores.

Moths are common, polyphagous pests in Murcia, Andalusia and Valencia, the main vegetable producing regions of Spain. In these areas, leaf-feeding noctuid lepidopterans, including *Spodoptera exigua* Hübner, *Autographa gamma* L. and *Chrysodeixis chalcites* Esper., and borer worms such as *Helicoverpa armigera* Hübner, *Ostrinia nubilalis* Hübner (Lepidoptera, Pyralidae) and *Gorthina xanthenes*, are gradually causing more damage and are becoming hard to control since fewer chemicals are available and problems of resistance are emerging (Torres-Vila *et al*., 2002).

The army worm, *Spodoptera littoralis* (Boisduval, 1833) (Lepidoptera: Noctuidae), is an important pest of cotton, tobacco and vegetable crops (tomato, pepper, lettuce, broccoli, cauliflower and artichoke). The larvae cause economically important damage since they feed on the leaves, fruits and buds from summer till the end of autumn in the field (or even year round when warm temperatures prevail during winter and spring) and year round in greenhouses (Rodríguez-Rodríguez *et al*., 1996; Téllez *et al*., 1996). Control methods include the use of agrochemicals such as pyrethroids, tebufe-

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Abbreviations used: AI (antifeedant indices); ppm (parts per million).

nocide and spinosad (organophosphates are gradually being banned), the use of *Bacillus thuringiensis* Berliner (Ramos-Gutiérrez *et al.,* 2004), baculovirus (Baculoviridae) (Murillo *et al*., 2006), and finally the use of pheromones (under development).

Plants are important sources of natural products with the potential to be developed as commercial active ingredients for pest control. Antifeedants have been proposed as an alternative to synthetic insecticides (Belles *et al*., 1985). Clerodane diterpenoids are well known insect antifeedants isolated from plants of the genera *Ajuga, Leonorus, Salvia, Scutellaria, Stachys* and *Teucrium* (family Labiatae) (Merrit and Ley, 1992; De la Torre *et al*., 1994; Rodríguez-Hanh *et al*., 1994; Bremner *et al*., 1998). The most potent neoclerodanes are jodrelin and scutalpin, which are isolated from *Scutellaria* (Anderson *et al*., 1989). The coumarins have been much less studied although they too have been cited as insect antifeedants (Addor, 1994; Stevenson *et al.,* 2003). In addition, a few papers document the antifeedant effects of flavones (Sharaby and Ammar, 1997) and other diterpenoids, for example those found in *Plectranthus* leaves (Wellsow *et al*., 2006).

This paper reports the antifeedant activity against *S. littoralis* of natural plant compounds belonging to different classes —clerodane, abietane, labdane and rosane diterpenoids, flavones and coumarins— with the aim of providing knowledge that may be of use in the development of new agrochemical products.

The phytochemicals used in this study are listed in Table 1 along with their plant sources.

Spodoptera littoralis was reared on a bean-based diet at 26 ± 1°C under a 16L:8D photoperiod. Larvae were originally collected several years ago from an alfalfa field in Totana (Province of Murcia); since then, this population has been reared in the lab to supply insects for research purposes.

Experiments were performed in 9 cm diameter Petri dishes each containing a glass fibre disc (diameter 2.1 cm) (Whatman GF/A). One hundred microlitres of a 50 mM sucrose solution plus 100 μL of a 100 ppm solution of the test compounds were applied to different discs (control discs received only the sucrose solution). A sixth instar larva was then added to each Petri dish. The discs (after the application of the products) were weighed before and after (18 h) the bioassay. Ten replicates were prepared for each compound assayed.

Following the methodology described by Simmonds *et al*. (1990), antifeedant indices (AI) were calculated according to the formula:

$$AI = [(C - T) / (C + T)] \times 100$$

where C = the weight (g) of the control disc consumed, and T = the weight (g) of the treated disc consumed. The AI ranged from –100 to +100. If a value of AI > 0 was returned, the compound was deemed to be an antifeedant; if a result of AI < 0 was returned, the compound was regarded as a phagostimulant. A mean AI was calculated for each assayed phytochemical. Comparisons between the eaten weight of the treated and control discs were made using the non-parametric Wilcoxon test.

The most active phytochemical was then assayed at different concentrations (4 doses, 2 replicates per dose, 100 larvae per replicate). Probit analysis (Robertson and Priesler, 1992) was then used to determine the doses with an antifeedant effect (AI ≥ 50) towards 10, 50 or 90% of the larvae.

Table 2 shows that the clerodane diterpenoids eriocephalin, salviacoccin and salvifarin, were active at 100 ppm, while the other compounds of this class had an AI value of around zero. The most active product (AI = 62.18) was eriocephalin. Overall, the abietane diterpenoids had less of an antifeedant effect, although aethiopinone and 2-oxocandesalvone A returned statistically significant results (AI = 29.82 and 23.02 respectively). In addition, larval feeding was reduced by the intake of 12-*O*-demethylcryptojaponol. As for the labdane and rosane diterpenoids tested, positive AI were obtained, although they were not much greater than those for the previously mentioned compounds (Table 2).

Eriocephalin, a compound isolated from *Teucrium eriocephalum* Wk, showed the greatest antifeedant effect. Table 3 shows the results of the probit analysis. The ED50 was 43 ppm, i.e., at this dose, an antifeedant effect (with AI ≥ 50) would be seen against 50% of larvae. European and Asian species of *Salvia* contain more abietane than clerodane diterpenoids (Rodríguez-Hanh *et al*., 1994). Aethiopinone is a strong anti-inflammatory substance (Hernández-Pérez *et al*., 1995) and possesses antimicrobial activity as well as cytotoxic activity towards cultured KB human carcinoma cells (Hernández-Pérez *et al*., 1999). Eriocephalin is known to be very active against *S. littoralis* (Simmonds *et al*., 1989; De la Torre *et al*., 1994). According to Geusken *et al*. (1983), substitutions in the decalin ring (4α,18-oxirane) and a β-substituted furan at C-12 are key structural features in this activity.

In contrast to the results reported here for *S. littoralis*, Ortego *et al*. (1995) reported that teucvin (a 19-nor

Table 1. Phytochemicals tested

***Class* / compound**	**Plant source[a]**	**Reference**
Clerodane diterpenoids		
Salviacoccin	*Salvia coccinea* Juss	Savona *et al.* (1982)
Salvifarin	*Salvia farinacea* Benth.	Eguren *et al*. (1984)
Eriocephalin	*Teucrium eriocephalum* Wk.	Fayos *et al.* (1979)
Isoeriocephalin	*Teucrium lanigerum* Lag.	Fernández-Gadea *et al.* (1984)
Teuflin	*Teucrium chamaedrys* L.	Fernández-Gadea *et al.* (1983)
Teucvin	*Teucrium chamaedrys* L	Rodríguez *et al.* (1984a)
Teuflidin	*Teucrium chamaedrys* L.	Fernández-Gadea *et al.* (1983)
Abietane diterpenoids		
Royleanone	*Salvia phlomoides* Asso.	Hueso-Rodríguez *et al.* (1983)
7α-Acetoxyroyleanone	*Salvia phlomoides* Asso.	
Taxodione	*Salvia phlomoides* Asso.	
Taxodone	*Salvia phlomoides* Asso.	
Crytojaponol	*Salvia phlomoides* Asso.	
12-O-Demethylcryptojaponol	*Salvia phlomoides* Asso.	
Horminone	*Salvia pratensis* L.	Hueso-Rodríguez and Rodríguez (1989)
12-O-Methylcandelabrone	*Salvia candelabrum* Boiss.	Mendes *et al.* (1989)
12-O-Methylcandesalvone A	*Salvia candelabrum* Boiss.	
12-O-Methylcandesalvone B 4-methyl ester	*Salvia candelabrum* Boiss.	
Aethiopinone	*Salvia aethiopis* L.	Rodríguez *et al.* (1984b)
2-Oxocandesalvone A 12-O-methyl ether	*Salvia palaestina* L.	Hussein *et al.* (1997)
Labdane diterpenoid		
Hispanolone	*Ballota hirsuta* Benth.	Savona *et al.* (1978)
Rosane diterpenoid		
Lagascatriol	*Sideritis angustifolia* Lag.	Martín-Panizo *et al.* (1974)
Flavones		
Salvigenin	*Sideritis serrata* Lag.	Rodríguez and Martín-Panizo 1979
Eupatorin	*Teucrium pseudochamaepitys* L.	Savona *et al.* (1979)
Cirsilineol	*Sideritis mugronensis* Borja	Rodríguez (1977)
5-Hydroxy-6,7,3’,4’-tetramethoxyflavone	*Sideritis mugronensis* Borja	
Gardenin D	*Sideritis mugronensis* Borja	
5,4’-Dihydroxy-6,7,8,3’-tetramethoxyflavone	*Sideritis mugronensis* Borja	
5-O-Demethylnobiletin	*Sideritis mugronensis* Borja	
Coumarins		
Isoimperatorin	*Cachrys trifida* Miller	Hernández and Rodríguez (1981)
Imperatorin	*Cachrys trifida* Miller	
Prantschimgin	*Cachrys trifida* Miller	
Bergapten	*Cachrys trifida* Miller	
Cnidilin	*Cachrys trifida* Miller	
Oxypeucedanin	*Cachrys trifida* Miller	
Xanthotoxin	*Cachrys trifida* Miller	
Prangol	*Cachrys trifida* Miller	

[a] All species are members of the family Labiatae, except for *Cachrys trifida* Miller which belongs to the family Umbelliferae.

Table 2. Antifeedant activity of phytochemicals (100 µL of a 100 ppm solution applied to 2.1 cm diam. discs) towards *Spodoptera littoralis*

Class / compound	AI (mean ± SEM)[a]	P[b]
Clerodane diterpenoids		
Salviacoccin	31.28 ± 17.669	0.0049*
Salvifarin	21.53 ± 19.179	0.1396
Eriocephalin	62.18 ± 11.338	0.0020*
Isoeriocephalin	0.39 ± 19.227	0.1797
Teuflin	4.14 ± 15.237	0.4043
Teucvin	0.25 ± 15.293	0.4219
Teuflidin	0.97 ± 22.587	0.1504
Abietane diterpenoids		
Royleanone	–37.20 ± 13.057	0.0420#
7α-Acetoxyroyleanone	6.11 ± 22.509	0.2539
Taxodione	–25.50 ± 18.276	0.1396
Taxodone	16.43 ± 13.922	0.0654
Crytojaponol	–10.90 ± 10.591	0.0967
12-O-Demethylcryptojaponol	33.42 ± 14.813	0.0645
Horminone	–21.00 ± 16.222	0.0820
12-O-Methylcandelabrone	10.39 ± 11.255	0.2305
12-O-Methylcandesalvone A	5.04 ± 18.275	0.3262
12-O-Methylcandesalvone B 4-methyl ester	20.92 ± 14.096	0.1650
Aethiopinone	29.82 ± 11.861	0.0273*
2-Oxocandesalvone A 12-O-methyl ether	23.02 ± 19.113	0.0186*
Labdane diterpenoid		
Hispanolone	11.64 ± 10.338	0.1132
Rosane diterpenoid		
Lagascatriol	23.43 ± 20.134	0.4043
Flavones		
Salvigenin	–8.75 ± 13.404	0.5332
Eupatorin	–5.19 ± 19.096	0.5332
Cirsilineol	15.46 ± 17.058	0.3633
5-Hydroxy-6,7,3’,4’-tetramethoxyflavone	–6.62 ± 15.876	0.4482
Gardenin D	–38.86 ± 24.109	0.0801
5,4’-Dihydroxy-6,7,8,3’-tetramethoxyflavone	38.31 ± 56.245	0.4043
5-O-Demethylnobiletin	–14.53 ± 15.156	0.3236
Coumarins		
Isoimperatorin	31.89 ± 22.921	0.0098*
Imperatorin	–31.38 ± 13.430	0.0420#
Prantschimgin	–33.79 ± 19.592	0.0039#
Bergapten	2.72 ± 17.996	0.4707
Cnidilin	8.86 ± 19.526	0.4707
Oxypeucedanin	41.92 ± 18.747	0.0137*
Xanthotoxin	43.99 ± 11.793	0.0186*
Prangol	26.78 ± 25.441	0.0195*

[a] Antifeedant index: $AI = [(C - T) / (C + T)] \times 100$; C = consumed weight of control disc, T = consumed weight of treated disc; $AI > 0$ = antifeedant activity, $A < 0$ = phagostimulant activity. [b] Probability for C – T (Wilcoxon test). * Statistically significant antifeedant activity ($P < 0.05$). # Statistically significant phagostimulant activity ($P < 0.05$).

Table 3. Regression analysis of eriocephalin antifeedant effect (AI ≥ 50) in *S. littoralis* larvae

ED10[a] (ppm)	95% Confidence limits	ED50[b] (ppm)	95% Confidence limits	ED90[c] (ppm)	95% Confidence limits
0.54	0.25-1.925	43	12-175	3469	1000- >10000

[a,b,c] Doses [ppm; solutions (100 µL) were applied to 2.1 cm diam. discs] with antifeedant effect (AI ≥ 50) towards 10%, 50% and 90% of larvae respectively.

neoclerodane) at 100 ppm caused a significant reduction in feeding in *Leptinotarsa decemlineata* larvae.

Salviacoccin and salvifarin have a saturated and α,β-unsaturated γ-lactone group respectively. According to Simmonds *et al*. (1996), this explains the activity of these compounds extracted from *Salvia.* These authors obtained an AI of 59 (at a dose of 100 ppm) for salviacoccin – a stronger antifeedant effect than that seen in the present experiment (AI = 31.28).

The flavones had no significant effect on the feeding behaviour of the larvae (Table 2).

The coumarins were active against *S. littoralis*. Oxypeucedanin and xanthotoxin gave AI values of over 40 (Table 2). A remarkable antifeedant activity was recorded for isoimperatorin (AI = 31.89) and prangol (AI = 26.78). The coumarins are a class of naturally occurring antifeedants that have been less studied than the neoclerodanes. Calcagno *et al*. (2002) reported a deterrent effect of xanthotoxin on *S. littoralis,* with synergistic effects in mixtures with imperatorin.

When the structures of active and inactive coumarins, such as isoimperatorin and imperatorin or xanthotoxin and bergapten are compared, substitution groups at different positions (C-4 or C-9) account for the differences in antifeedant activity (Table 1).

Some papers report the medicinal potential of furocoumarins such as imperatorin, isoimperatorin, oxypeudanin and prantschimgin (Abad *et al*., 2001; Hyuncheol *et al*., 2002).

In summary, the active diterpenoids isolated from species of *Teucrium* and *Salvia* (Labiatae) and a number of coumarins from *Cachrys trifida* (Umbelliferae), appear to have antifeedant potential with respect to *S. littoralis*. More research is required in this area to help find alternatives to banned pesticides.

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